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^{129}I and ^{137}Cs in Groundwater in the Vicinity of Fukushima Dai-ichi Nuclear Power Plant

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Abstract

This paper reports iodine (^{127}I and ^{129}I) and cesium (^{137}Cs) isotope concentrations in groundwater of confined and unconfined aquifers in the vicinity of the Fukushima Dai-ichi nuclear power plant (FDNPP). ^{127}I and ^{129}I concentrations range from 2-13 μg/L and 5×10^{7}-8×10^{10} atom/L respectively, resulting in $^{129}I/^{127}I$ atomic ratios from 5×10^{-9} to 2×10^{-6}. In all samples, ^{137}Cs concentrations were below detection limit. The deep-sealed groundwater from the confined aquifer did not contain significant quantities of Fukushima-derived ^{129}I compared to the groundwater in the unconfined aquifer. The minimal $^{129}I/^{137}Cs$ activity ratios in the groundwater are more than 2-500 times higher than the FDNPP source ratio. These data can be explained by rainwater infiltrating through the surface soils, with the more water-soluble ^{129}I preferentially extracted into the aqueous phase and the ^{137}Cs preferentially retained in the soil.

INTRODUCTION

Following the accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), radionuclide deposition resulted in serious radioactive contamination in the area near FDNPP, extending to a large area of eastern Japan. Numerous investigations on Fukushima-derived radionculides in the various environments have been published recently. These include studies of aerosols, rainwater, seawater, soil, plants, animals, etc. from the local and worldwide environments. Among these studies some have focused on the migration of radiocesium (^{134}Cs and ^{137}Cs) and radioiodine (^{131}I) in the soil layer (Ohta et al., 2012; Tanaka et al., 2012; Saito et al., 2014). It has been argued that the migration rates of ^{137}Cs are so low that contamination of groundwater by ^{137}Cs is not likely to occur in rainwater infiltrating into the surface soil after the Fukushima accident (Ohta et al., 2012). However there are very few direct measurements of radioiodine and radiocesium in groundwater samples. For this purpose, this study determined ^{129}I and ^{137}Cs concentrations in five groundwater samples in addition to one rainwater sample near the FDNPP to assess the levels of Fukushima-derived radionuclides in confined and unconfined aquifers.

MATERIALS AND METHODS

Five groundwater samples were collected in the vicinity of the FDNPP on August 28, 2014 (Fig. 1). Sample Nos.1, 2 and 4 were collected from residential wells with the water surface 1.2 m, 1.8 m and 1.1 m below the ground surface, respectively. Sample No. 3 was also collected from a residential well, but water was overflowing from the well following the earthquake on March 11, 2011. Water in Site 5 was seeping from a channel in a gentle escarp, which is ~1.5 m below surface. Long-term monitoring of water level in Well Nos. 1, 2 and 4 shows rapid response to rainfall, indicating that rainfall rapidly penetrates the soil to these near surface aquifers. As described below, Marui (2015) also concluded that shallow groundwater within the FDNPP is generated by recent rainfall. As a result, an additional rainwater sample (No. 6) was contemporarily collected from Namie, ~7 km north of the FDNPP, which is considered representative of rainfall that had recently contributed to these aquifers.

Groundwater samples were first measured for pH, conductivity, salinity and oxidation-reduction potential (ORP). Aliquots of water samples (~100 mL) were filtered through a 0.45 μm membrane. The ^{127}I and ^{137}Cs in the filtered water were determined using inductively coupled plasma mass spectrometer and gamma spectrometer in the Technical University of Denmark. ^{129}I concentration was measured by chemical extraction of iodine from the water combined with determination using a 5 MV tandem accelerator mass spectrometer (AMS) in the Scottish Universities Environmental Research Center. Detailed experimental procedures have been previously described in Xu et al. (2013; 2015).

RESULTS AND DISCUSSION

Analytical results including $^{127,129}I$ concentrations and $^{129}I/^{127}I$ ratios together with water chemical and physical compositions are listed in Supplementary Table S1. Data of pH and ORP suggest two groups: one with low pH (6.23-6.87) and high ORP (192-298 mV), the other with high pH (8.5) and low ORP (20 mV). The electrical conductivity (7-70 mS/m) and salinity (0.01-0.03 %) overlap between the two groups. These groups are consistent with groundwater from shallow and deep strata below the FDNPP (Marui, 2015) which also indicate distinctive grouping for pH and conductivity for the deep and shallow aquifers. Figure 2 illustrates the relationship between pH and electrical conductivity of groundwater in this study and Marui (2015), showing that samples 1, 2, 4 and 5 from this study are consistent with shallow waters and sample 3 consistent with deeper water.

^{137}Cs concentrations are below the detection limit (<0.5 Bq/L) in all samples. The measured ^{127}I and ^{129}I concentrations vary from 2 μg/L to 13 μg/L and from 5×10^{7} to 8×10^{10} atom/L respectively, resulting in $^{129}I/^{127}I$ atomic ratios from 5×10^{-9} to 2×10^{-6}. Figure 3 shows the relationship between ^{127}I and ^{129}I. The variation in ^{127}I is relatively small, reflecting variations in soil geochemistry, therefore the large variations in the $^{129}I/^{127}I$ ratio are mainly controlled by ^{129}I in the groundwater system.

Hydrochemical and hydrological features of groundwater

The most striking feature in this study is that sample No. 3 has higher pH, lower ORP, ^{129}I, $^{129}I/^{127}I$ than other samples. This suggests that the source of groundwater in sample No. 3 is most likely different from the other samples.

Groundwater can hydrologically originate from the confined and unconfined aquifers. Indeed, all shallow groundwater in this study is generally catalogued into the unconfined aquifer. Marui (2015) conducted several boring holes within the FDNPP for geomorphological and geological investigations. The shallower ground water (the mid-sized sandstone stratum I and the alternate strata stratum III) have lower pH values of 6.1-6.9, and the deeper water from coarse and fine sandstone stratum IV has higher pH of 7.5-9.0. This chemical composition is consistent with the two groups of pH values in our study (Fig. 2), with sample No. 3 being from the deeper source. The deeper aquifer might be confined, otherwise it would not be under sufficient pressure to rise to the surface and overflow the top of Well No. 3. The shallow aquifer is probably in the Quaternary terrace deposits, which are estimated to be 5-10 m deep at the well sites (far deeper than the 1-2 m below surface of the top of the water table).

Within the FDNPP site and its adjacent areas, the strata I-IV generally eastwardly incline with the dip angle ~30º (Marui, 2015). Extrapolating the cross section, at our No. 3 the stratum IV would be about 10 m below the surface without any of the younger sandstone strata (I-III) above it. The level of water table in the Well No. 3 used to be similar to others. However, it was elevated and water was overflowing to surface after the earthquake. It is suggested that the earthquake might have caused a blockage of the eastward water flow underneath the well, but also resulted in fractures from which groundwater in the stratum IV can flow upward. Therefore, these observations support sample No. 3 being considered as a representative of deep-sealed groundwater from the confined aquifer, whereas groundwater in other sites originated from the upper unconfined aquifer.

Radionuclides in groundwater in the confined aquifer

Sample No. 3 has the regional lowest ^{127}I and ^{129}I concentration of 2 μg/L and 4.6×10^{7} atom/L respectively. They were obviously lower than those (4 μg/L and 2.4×10^{8} atom/L) in the contemporary rainwater (No. 6) collected nearby. The ^{129}I concentration is also clearly lower than those in rainwater from Fukushima city prior to the Fukushima accident (1.0-2.8×10^8 atom/L between November 2010 and February 2011 (Xu et al., 2013). The $^{129}I/^{127}I$ ratio of 4.5×10^{-9} is not only lower than that (1.3×10^{-8}) in the contemporary rainwater, but also much lower than those in rainwater from Fukushima city since the Fukushima accident (Xu et al., 2013). Furthermore, the $^{129}I/^{127}I$ ratio in this groundwater is also lower than those determined in water samples from the background area at Chiba in 1983 (1×10^{-8}, Muramatsu et al., 1986), in the atmospheric fallout in Tokyo from 1963 to 1980 ($1\text{-}2\times10^{-8}$, Toyama et al., 2012), and in surface soils in Fukushima before the accident ($<3\times10^{-8}$, Matsuzaki et al., 2007). Therefore, it can be concluded that the deep-sealed groundwater in the confined aquifer in the vicinity of FDNPP has not been contaminated by Fukushima-derived radionuclides.

However, the ^{129}I concentration here is significantly higher than previous estimates of 10^3 to 10^4 atom/L for groundwater prior to nuclear weapons testing (Rao and Fehn, 1999; Alvarado Quiroz et al., 2002). Previous studies have indicated that brackish to saline waters in granites can have ^{129}I concentrations between 2×10^6 and 3.4×10^8 atom/L attributed to leaching of ^{129}I produced by spontaneous fission of ^{238}U in granites (Moran et al., 1995; Kotzer et al., 1998). Local geological evidence also supports that ^{129}I in the confined aquifer is most likely derived from spontaneous fission of ^{238}U enriched in the basement Cretaceous granites beneath the FDNPP (Tsutsumi et al., 2010).

Radionuclides in groundwater in the unconfined aquifer

Groundwater samples in other sites (Nos. 1, 2, 4 and 5) have ^{127}I and ^{129}I ranging in 4-13 μg/L and $(3\text{-}836)\times10^8$ atom/L, respectively. In contrast to the narrow range of ^{127}I, a large range in $^{129}I/^{127}I$ from 1.6×10^{-8} to 2.1×10^{-6}, over two orders of magnitude, is found within the region near the FDNPP. The ^{127}I, ^{129}I and $^{129}I/^{127}I$ values are significantly higher than those in the contemporary rain (No.6). The highest ^{129}I and ^{129}I/^{127}I values in these groundwater samples are similar to those in rainwater from Fukushima city in June 2011 (Xu et al., 2013) and comparable with those in the top surface soils within 5 km from the FDNPP (~3×10^{-6}, Miyake et al., 2015). The ^{129}I dataset indicates that there are variable but significant amounts of Fukushima-derived ^{129}I in groundwater in the unconfined aquifer near the FDNPP. The high detection limit for ^{137}Cs in these measurements does not allow quantification of activity concentrations in these groundwater samples. However, assuming that the detection limit is the maximal ^{137}Cs value in groundwater, the minimal ^{129}I/^{137}Cs activity ratios in groundwater (>9-2000×10^{-7} Bq/Bq) are >2-500 times higher than the Fukushima-derived source value (~4×10^{-7}, Tumey et al., 2013; Xu et al., 2015). It is also significantly higher than those in the surface soils distributed within 5 km from the FDNPP (~5×10^{-7}, Miyake et al., 2015; Fujiwara et al., 2012).

Questions then arise regarding the mechanism for Fukushima-derived radioiodine migration into the groundwater in the unconfined aquifer. Radionuclides released during the Fukushima accident have been deposited on the land near the FDNPP and a large area of eastern Japan. The fact that ^{129}I/^{127}I ratio is similar for water and topmost section of surface soil near the FDNPP suggests that ^{127}I and ^{129}I in the water is derived from the surface soil. The FDNPP is located on Quaternary alluvial deposits composed of clay and sand. The middle layer (20-30 m above sea level) consists of sandy loam while the overlying (0-20 m above sea level) are mainly of sandstone (Saeki, 1967; Marui, 2015). Such local lithology suggests that the migration of ^{129}I and ^{137}Cs into a deeper soil layer (>5 cm) is restricted due to their strong affinities for humic substances and clay minerals, respectively (Ohta et al., 2012; Tanaka et al., 2012; Saito et al., 2014). Nevertheless, water-leaching experiments on soil have shown that less than 1 % of ^{137}Cs and about 10 % of ^{131}I can be dissolved into the aqueous phase at any pH (Tanaka et al., 2012). Sequential extraction experiments have also indicated that 5-15 % ^{137}Cs and 42-61 % ^{129}I in aerosols were water-soluble (Xu et al., 2015). These observations confirm that iodine is much more water-soluble than cesium, which can result in large fractionation between iodine and cesium. It is noted that cesium and iodine may also be carried by nano- or micrometer scale particles which may also be transported by water from the surface to the relatively shallow aquifers. However, the below detection limit concentrations of ^{137}Cs observed indicate that this is not a significant ^{137}Cs transport mechanism for these samples. The large range in ^{129}I/^{127}I in groundwater, over two orders of magnitude, within the region near the FDNPP may imply that Fukushima-derived ^{129}I and natural ^{127}I are transported separately across the region, presumably related to differences in chemical form and time since deposition.

CONCLUSIONS

Fukushima-derived radionuclides are not observed in groundwater in the confined aquifer near the FDNPP, however, they have been found in the unconfined aquifer. Rain infiltration extracts water-soluble ^{129}I and ^{137}Cs from contaminated soils. As ^{129}I is much more extractable than ^{137}Cs, large fractionation between ^{129}I and ^{137}Cs has occurred in the aqueous phase. The variations in Fukushima-derived radionuclide concentrations in groundwater at different locations may be of benefit in understanding the current hydrology of the area surrounding the FDNPP site.

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Figure captions

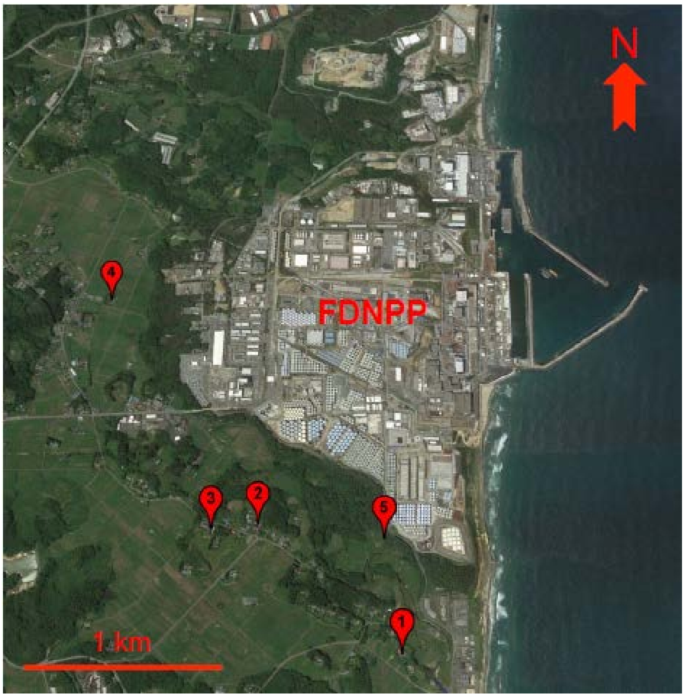

Figure 1. Map showing the FDNPP and sampling locations of groundwater. Map data © 2015 Google, image © 2015 TerraMetrics.

Figure 2

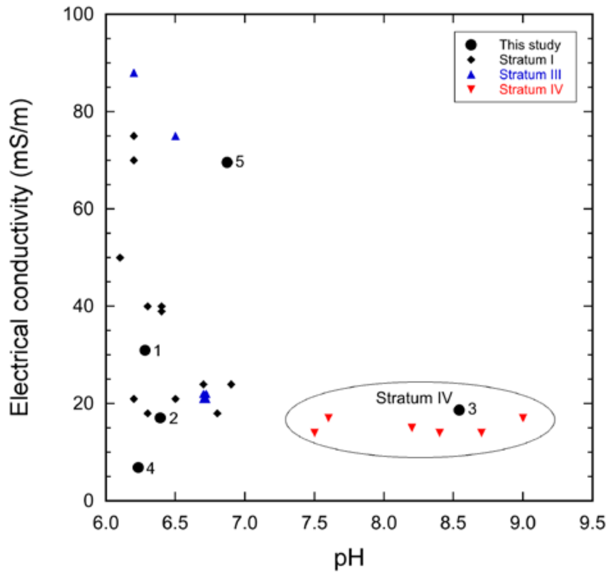

Figure 2. Relationships between pH and electrical conductivity of groundwater for different strata beneath the FDNPP (Marui, 2015), and for the samples in this study. Strata I and III are shallow (<10 m below the surface at FDNPP) and stratum IV deeper (20-30 m below the surface at FDNPP). Samples 1, 2, 4 and 5 from this study show characteristics consistent with the shallow ground waters from Marui (2015), and sample 3 with the deeper ground waters.

Figure 3

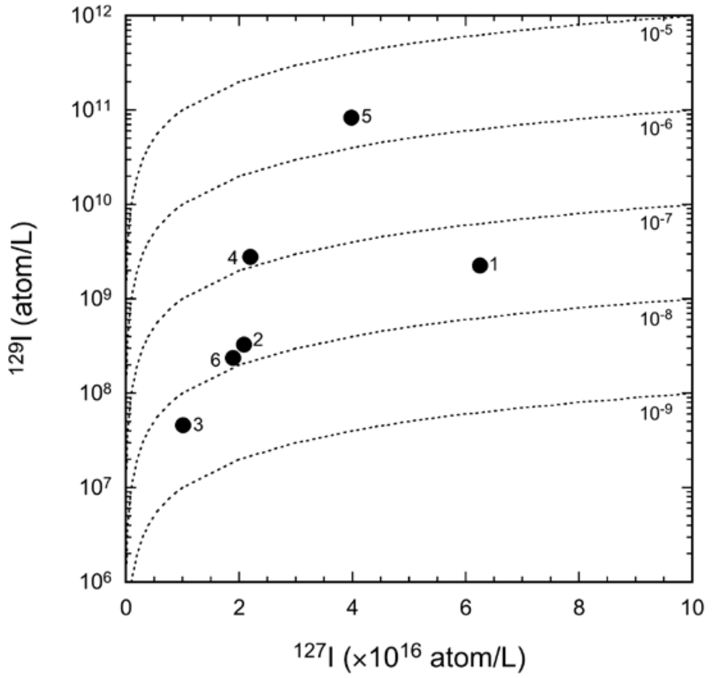

Figure 3. Correlation between ^{127}I and ^{129}I in water samples near the FDNPP. The dotted lines denote ^{129}I/^{127}I atomic ratios